Electronic Supplementary Information

Three-dimensional Graphene/polypyrrole hybrid Electrochemical Actuator

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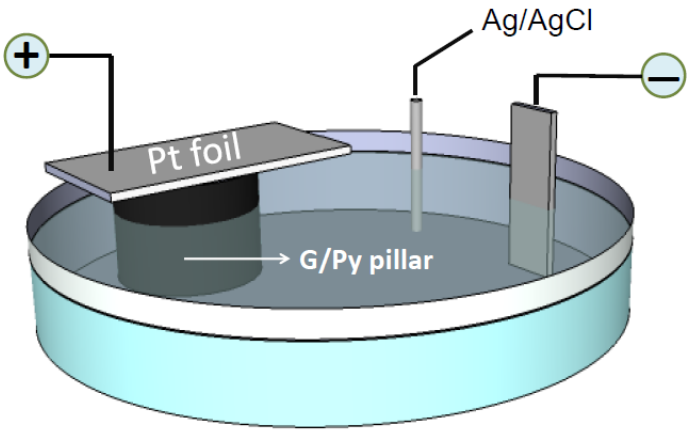
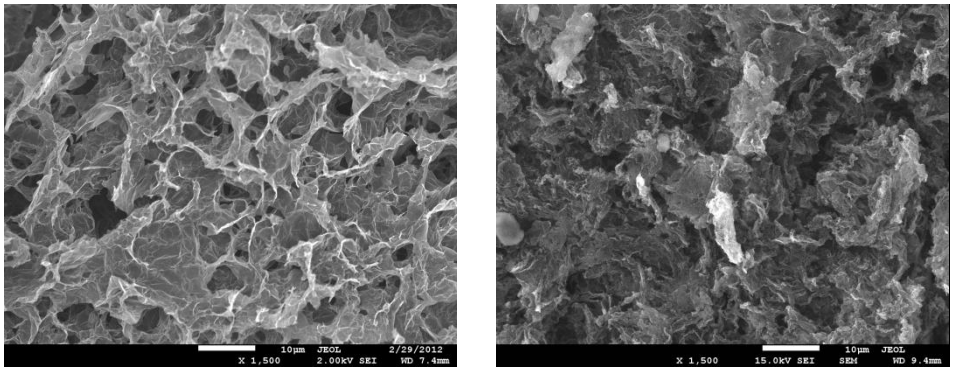


Fig. S1 A schematic diagram showing the electropolymerization of pyrrole containing in the 3D graphene structure.



(a) (b)

Fig. S2 SEM images of the 3D graphene/PPy prepared by directly electropolymerization of the preformed pyrrole-containing 3D graphene without any additional Py monomer (a) and synthesized by electropolymerization of additional Py monomer onto pure 3D graphene framework.

The mixture of GO with Py monomer prior to the hydrothermal process is crucial for formation of 3D uniform graphene/PPy structure. As shown in Figure S2, after undergoing the hydrothermal process, the aforehand mixed Py monomer will uniformly polymerize along the whole surfaces of graphene sheets and no aggregated particles are observed within the 3D graphene structure (Figure S2a). In contrast, the pure 3D graphene will lose porosity and the serious aggregation of PPy appears around the graphene sheets after electropolymerization of additional Py monomer onto pure 3D graphene framework (Figure S2b).

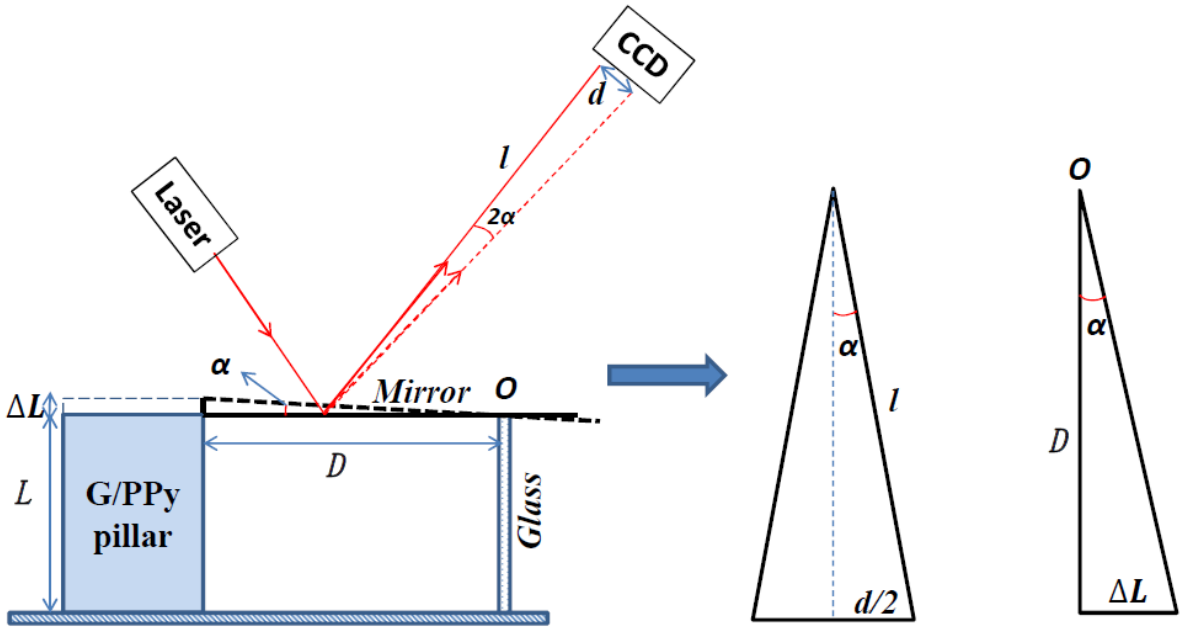


Fig. S3 Scheme and the relative parameters for calculation of strain response of 3D G/PPy actuator.

Fig. S3 exhibits the relative parameters of 3D G/PPy actuator for strain response calculation. Each parameter is defined as follows:

L: The original height of 3D G/PPy actuator.

ΔL: The variational displacement in height of 3D G/PPy actuator.

D: The original distance between the fulcrum of glass and 3D G/PPy pillar.

d: The displacement measured by CCD.

l: The distance between the laser point and CCD.

α: Rotation angle of the mirror.

According to the reflection law, once fixed the incident ray, the mirror rotates an angle of α, while the reflection ray will rotate an angle of 2α.

Obviously,

$$\sin\alpha = d/2l$$

$$\tan\alpha = \Delta L/D$$

In current case, l (*ca.* 0.9m–1m) >> d (*ca.* 0.1mm–1mm),

$$\alpha \approx \sin\alpha \approx \tan\alpha$$

It is deduced that

$$\Delta L = Dd/2l$$

Thus, the strain of the actuation response is

$$\text{Strain (\%)} = \Delta L/L = Dd/2Ll$$

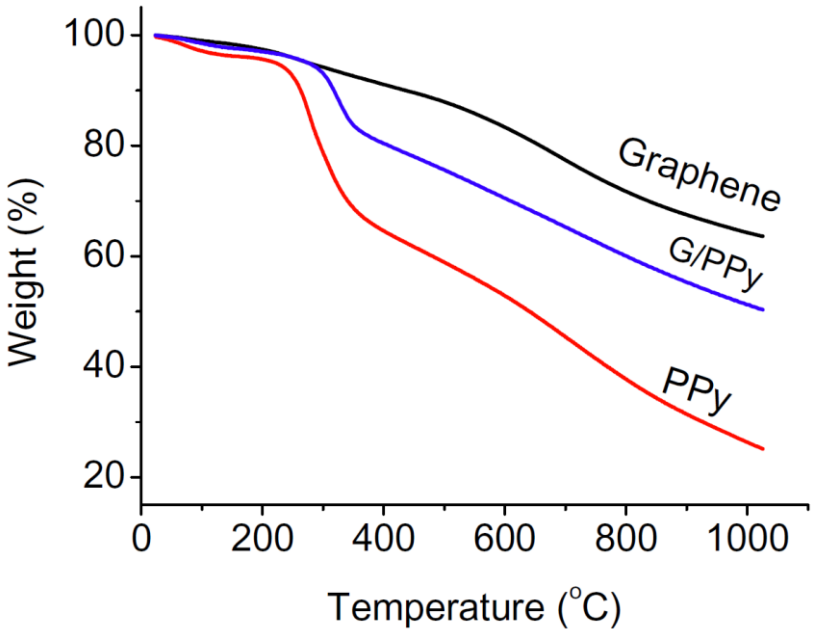


Fig. S4 TGA curves of hydrothermally synthesized 3D graphene, electropolymerized PPy and the as-prepared 3D G/PPy, respectively.

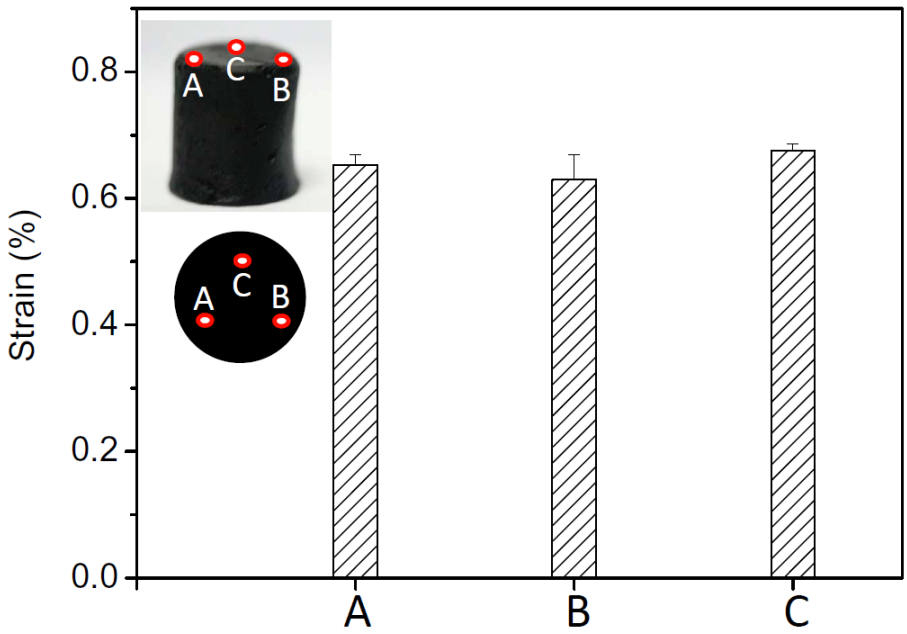


Fig. S5 Comparison of the strain response of three points on 3D G/PPy pillar under applied square wave potential of ± 0.8 V with a cycle time of 50 s. Insets show the side and top view of the measured points on the sample.